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Backscattering properties of gold nanoshells: quantitative analysis and optimization for biological imaging

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Abstract

In this article, we investigate the dependence of the backscattering properties of gold nanoshells on the core material, core radius, and shell thickness by using Mie theory of a coated sphere. The size-dependent and dispersive dielectric function of metal nanoparticles permits to consider the localized surface plasmon resonance of gold nanoshells with the classical Mie theory. Our results show that gold nanoshells exhibit resonant backscattering feature due to the resonance of the free electrons in gold shells at a specific incident wavelength. The resonance peak intensity depends on the core material and the dimensions of core and shell, and the resonance wavelength can be tuned to the near-infrared region where biological tissues possess high transmissivity. We obtain the maximum backscattering and the corresponding optimal core radii and shell thicknesses of gold nanoshells at four typical wavelengths (830, 840, 900, and 1310 nm) frequently-used in biological imaging. It is found that silica core-gold nanoshells have maximum volume backscattering coefficient (7.01 μm^{-1}) at wavelength of 830 nm when the core radius is 54.2 nm and the shell thickness is 10.1 nm. For longer wavelength, the optimal core radius is larger while the optimal shell thickness is thinner, and the maximum volume backscattering coefficient become small. If the core material is changed from silica to vacuum, the gold nanoshells have bigger maximum volume backscattering coefficient with larger core radius and thinner shell thickness. The optimized gold nanoshells can be used as the ideal contrast agents for biological imaging.

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Keywords: Gold nanoshells; Backscattering; Biological imaging; Contrast agent; Optimization

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1. Introduction

Gold nanoshells, a novel type of composite spherical nanoparticle consisting of a spherical dielectric core covered by an ultrathin gold shell with uniform thickness, are able to strongly absorb and/or scatter light at a specific wavelength within the visible and near-infrared region due to the localized surface plasmon resonance. A localized surface plasmon is a collective spatial oscillation of the free electrons in a metal nanoparticle; it can be directly excited by light and resonate at a specific wavelength. By varying the dimension and composition of each layer of the nanoshell structure, the resonance wavelength can be tuned to any wavelength desired across a large region of the visible and near-infrared, and the extent of light absorption and scattering can also be tuned [1-7]. The tunable optical properties of gold nanoshells, coupled with high chemical stability and good biocompatibility, make them have great potential in biomedical applications.

In recent years, gold nanoshells have been used as therapeutic agents for photothermal cancer therapy based on their strong absorption in the near-infrared region where optical transmission through tissue is optimal [8-14]. In addition to their potential applications as therapeutic agents, gold nanoshells have also been used as contrast agents for biological imaging with optical coherence tomography (OCT) due to their strong backscattering in the near-infrared region [15-19]. Loo et al. [15] were the first to demonstrate the feasibility of using gold nanoshells as contrast agents for biological imaging with OCT. Agrawal et al. [16] investigated the influence of geometry and concentration of gold nanoshells on OCT signal enhancement in turbid media by in vitro OCT measurements. Their experimental results indicated that gold nanoshells with high backscattering are the effective contrast agents for OCT imaging. Zagaynova et al. [17] demonstrated the efficiency of gold nanoshells as contrast agents for OCT imaging of agar biotissue phantoms and rabbit skin in vivo. Kah et al. [18,19] studied the dependence of concentration of gold nanoshells on OCT signal enhancement. They found that too high a concentration of gold nanoshells only enhances the OCT signal near the tissue surface, while significantly attenuating the signal deeper into the tissue. Since OCT detects backscattered light originating from index mismatches in tissue, gold nanoshells with higher backscattering at the OCT source wavelength can potentially be important in enhancing OCT signals from the tissue. Therefore, strong backscattering of gold nanoshells is essential for biological imaging with OCT. To the best of our knowledge, no systematic investigation has ever been performed for the backscattering properties of gold nanoshells and no optimization has ever been carried out for the dimensions of gold nanoshells by maximizing the backscattering. Therefore, the goal of this study is to fill this gap by providing quantitative analysis and optimization for the backscattering properties of gold nanoshells.

This paper is organized as follows. Section 2 introduces the calculation of the backscattering properties of gold nanoshells by using Mie theory of a coated sphere with size-dependent dielectric function of metal nanoparticles. In Section 3, we investigate the effects of the core material and particle size on the backscattering properties of gold nanoshells, and obtain the maximum backscattering and the corresponding optimal core radii and shell thicknesses for silica and hollow core gold nanoshells at four typical wavelengths (830, 840, 900, and 1310 nm) commonly-used in biological imaging with OCT. Section 4 is devoted to the conclusion.

2. Theory

2.1. Backscattering of gold nanoshells

The geometry of the gold nanoshell studied in this paper is shown in Fig. 1. The backscattering properties of an individual gold nanoshell can be described by backscattering cross section (C_{back}) which represents an equivalent cross sectional area of the particle that contributes to backscattering of the incident light. The C_{back} of an individual gold nanoshell can be calculated by using the Mie theory of a coated sphere [20]

$$C_{back} = \frac{1}{4k^2}\left|\sum_{n=1}^{\infty}(2n+1)(-1)^n\left(a_n - b_n\right)\right|^2, \tag{1}$$

where k is the wavenumber in the surrounding medium, and a_n and b_n are the scattering coefficients which can be calculated by employing the algorithm proposed by Bohren and Huffman [20].

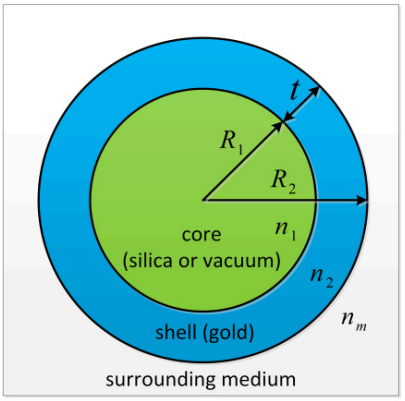

Fig. 1. Geometry of gold nanoshell. R_1, R_2, and t are the core radius, shell radius, and shell thickness, respectively. n_1, n_2, and n_m are the refractive indices of the core, shell, and surrounding medium, respectively.

The backscattering properties of an ensemble of gold nanoshells can be described by backscattering coefficient (μ_{back}) which represents the total backscattering cross section of gold nanoshells within unit volume. If multiple scattering is negligible and the gold nanoshell ensemble is monodisperse (i.e., the gold nanoshells have the same dimensions), the μ_{back} is given by

$$\mu_{back} = NC_{back} = f\frac{C_{back}}{V_p}, \tag{2}$$

where N is the particle number density (i.e., the number of particles per unit volume), f is the volume fraction of particles (i.e., the volume of particles per unit volume), V_p is the volume of an individual particle. In this paper, we assumed that gold nanoshell ensembles with different dimensions have the same volume fraction in order to the compare the backscattering properties fairly. Therefore, we defined a new quantity, which is the backscattering cross section per unit particle volume, called volume backscattering coefficient (α_{back}) to compare the backscattering properties of gold nanoshells ensembles. The α_{back} is given by

$$\alpha_{back} = \frac{C_{back}}{V_p}. \tag{3}$$

We developed a computer code for the calculation of α_{back}. The required parameters for the code are the vacuum wavelength of incident light λ, core radius R_1, shell thickness t, and refractive indices of the core, shell, and surrounding medium, n_1, n_2, and n_m, respectively. In our calculations, the core material was considered to be silica with refractive index given by Malitson [21] or vacuum with refractive index of 1.0, and the surrounding medium was considered to be biological tissue with refractive index of 1.4 [22]. The refractive index of the gold shell (n_2) can be obtained from the dielectric function ε of the gold shell by $n_2 = \varepsilon^{1/2}$. The calculation of the dielectric function for the gold shell is described in the next subsection.

2.2. Size-dependent dielectric function

For bulk metals, the dielectric function is dependent on the angle frequency of the incident light (ω), and it is the sum of the contributions from the bound and free electrons in the metals,

$$\varepsilon_{bulk}(\omega) = \varepsilon_{bound}(\omega) + \varepsilon_{free}(\omega), \tag{4}$$

where ε_{bound} and ε_{free} originate from the interband and intraband transitions, respectively. The contributions from the free electrons can be described by the Drude model [20],

$$\varepsilon_{free}(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\gamma_{bulk}}, \tag{5}$$

where ω_p is the plasma frequency, and $\gamma_{bulk}=v_f/l_\infty$ is the bulk collision frequency dependent on the Fermi velocity v_f and the mean free path of the free electrons l_∞.

The dielectric function of a bulk metal is independent on size. However, when the light interacts with the small metal nanoparticles of the size smaller than the mean free path of the free electrons, the dielectric function of the nanoparticles deviates from the bulk value. The bulk collision frequency increases because the collision of the free electrons with the particle surface becomes important as an additional relaxation process and cannot be neglected [23]. Therefore, when the size-dependent free electron surface scattering become important, the bulk collision frequency should be modified as follows,

$$\gamma = \gamma_{bulk} + Av_f / L_{eff}, \tag{6}$$

where A is a dimensionless parameter which is usually assumed to be close to 1, and L_{eff} is the effective mean free path of the free electrons dependent on the dimensions of the metal nanoparticles. Thus, for small metal nanoparticles, the dielectric function should be modified to account for the surface scattering of the free electrons, which is expressed as

$$\begin{aligned}\varepsilon(\omega, L_{eff}) &= \varepsilon_{bound}(\omega) + \varepsilon_{free}^{nano}(\omega, L_{eff}) = [\varepsilon_{bulk}(\omega) - \varepsilon_{free}(\omega)] + \varepsilon_{free}^{nano}(\omega, L_{eff}) \\ &= \varepsilon_{bulk}(\omega) + \frac{\omega_p^2}{\omega^2 + i\omega v_f / l_\infty} - \frac{\omega_p^2}{\omega^2 + i\omega(v_f / l_\infty + Av_f / L_{eff})}\end{aligned}. \tag{7}$$

For gold nanoshells, $\hbar\omega_p$ = 9.03 eV [24], $v_f = 1.40\times10^{15}$ nm/s [3], l_∞ = 42 nm [3], A = 1 [3], $L_{eff} = t$ [3], and the values of the bulk dielectric function ε_{bulk} were obtained from the literature reported by Johnson and Christy [25].

3. Results and discussion

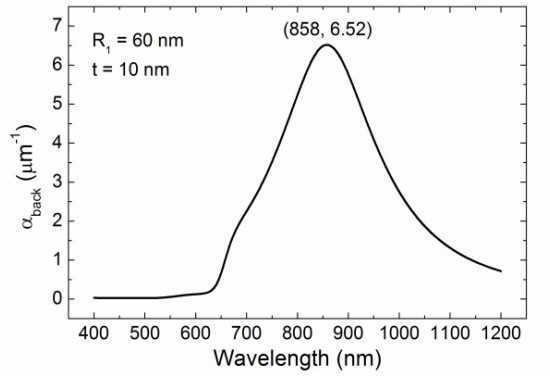

Fig. 2. Calculated spectrum of volume backscattering coefficient (α_{back}) for silica core-gold nanoshells. The core radius (R_1) and shell thickness (t) are 60 and 10 nm, respectively.

Gold nanoshells can be used as optical contrast agents for biological imaging applications due to their strong and tunable backscattering in the near-infrared wavelength region where biological tissues possess high transmissivity. Therefore, it is very important to perform a comprehensive investigation of the backscattering properties of gold nanoshells. Fig. 2 shows the spectrum of volume backscattering coefficient for silica core-gold nanoshells with the 60-nm core radius and 10-nm shell thickness. It is seen that there is a resonance peak presented in the spectrum at the wavelength of 858 nm, and the peak intensity is 6.52 μm^{-1}. In other words, gold nanoshells exhibit resonant backscattering feature which is attributable to the resonance of the free electrons in gold shells while interacting with light. In the remainder of this section, we provided systematic numerical simulation on the spectra of volume backscattering coefficient of gold nanoshells in order to investigate the effects of the dielectric core and particle size on the position and intensity of the resonance peak. In the end, we optimized the dimensions of gold nanoshells to obtain the strongest backscattering at several typical wavelengths which is frequently-used in biological imaging.

3.1. Effect of core material

Advance in chemical and physical synthesis techniques of nanoparticles have made the fabrication of gold nanoshells with various cores possible. Here we selected two common gold nanoshells (silica core-gold nanoshells and hollow core-gold nanoshells) as the objects of investigation in order to analyze the effect of the core material on the position and intensity of the backscattering resonance peak. The spectra of volume backscattering coefficient for silica and hollow core-gold nanoshells with the 60-nm core radius and 10-nm shell thickness is shown in Fig. 3. When the core material is changed from silica to vacuum, the resonance wavelength decreases from 858 to 826 nm, and the peak intensity increases from 6.52 to 6.98 μm^{-1}. Thus hollow core-gold nanoshells have stronger backscattering resonance at smaller wavelength as compared to silica core-gold nanoshells having the same dimensions.

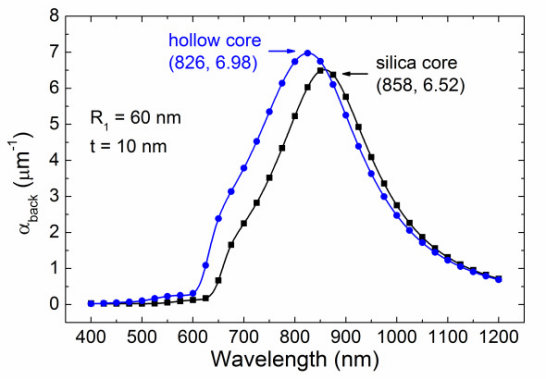

Fig. 3. Calculated spectra of volume backscattering coefficient (α_{back}) for gold nanoshells with silica (square) and hollow (circle) cores. The core radius (R_1) and shell thickness (t) are 60 and 10 nm, respectively.

3.2. Effect of particle size

Using current synthesis techniques, gold nanoshells of a wide variety of core and shell sizes can easily be fabricated. We thus preformed quantitative analysis for the effects of the dimensions of gold nanoshells on their backscattering resonance peak. Fig. 4 summarizes the dependence of the backscattering resonance peak of gold nanoshells on the core radius. As shown in Fig. 4(a), as the core radius is increased from 20 to 100 nm, the resonance peak red-shifts near-linearly from 611 to 1115 nm for silica core-gold nanoshells and from 598 to 1083 nm for hollow core-gold nanoshells. The silica core-gold nanoshells possess higher resonance wavelength when compared with the hollow core-gold nanoshells of the same dimensions, and the difference between the resonance wavelengths becomes a little larger with the increase of core radius. Fig. 4(b) shows that the peak intensity increases sharply first, then reaches a maximum value, and finally decreases with the increase of core radius. Silica core-gold

nanoshells (hollow core-gold nanoshells) have maximum peak value of 9.23 μm^{-1} (10.41 μm^{-1}) at the resonance wavelength of 684 nm (674 nm) when the core radius is 33 nm (35 nm). It is seen that the resonance peak intensity of silica core-gold nanoshells is weaker than that of hollow core-gold nanoshells of the same dimensions.

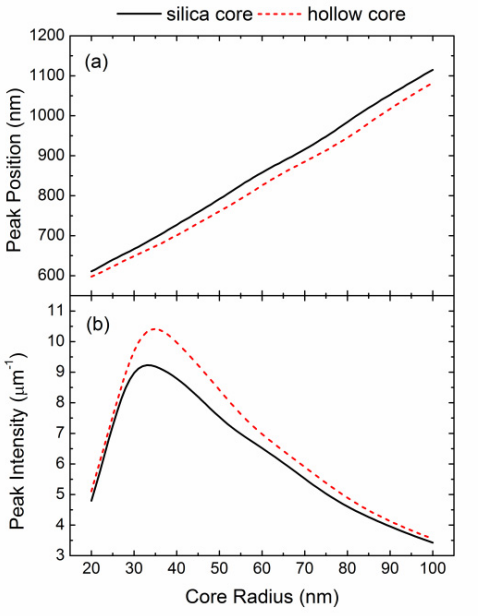

Fig. 4. (a) Position and (b) intensity of the backscattering resonance peak of silica (solid) and hollow (dash) core-gold nanoshells as a function of core radius varied from 20 to 100 nm. The shell thickness is fixed at 10 nm.

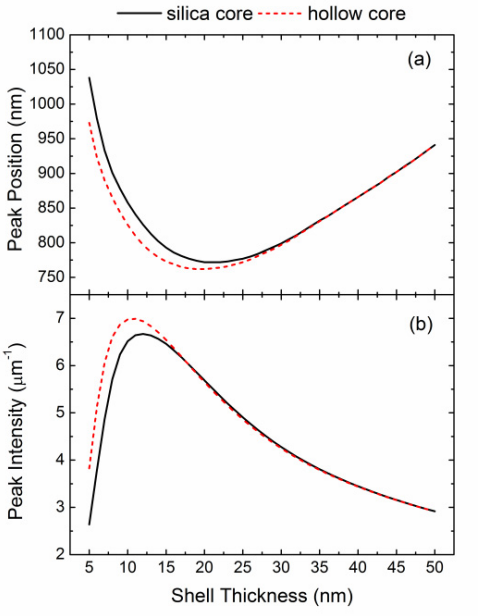

Fig. 5. (a) Position and (b) intensity of the backscattering resonance peak of silica (solid) and hollow (dash) core-gold nanoshells as a function of shell thickness varied from 5 to 50 nm. The core radius is fixed at 60 nm.

The dependence of the backscattering resonance peak of gold nanoshells on the shell thickness is displayed in Fig. 5. As the shell thickness is increased from 5 to 50 nm, the resonance wavelength decreases sharply first, then reaches minimum, and finally increases [Fig. 5(a)], while the peak intensity increases first, then reaches maximum, and finally decreases [Fig. 5(b)]. Silica core-gold nanoshells (hollow core-gold nanoshells) have maximum peak value of 6.67 μm^{-1} (6.99 μm^{-1}) at the resonance wavelength of 826 nm (810 nm) when the shell thickness is 12 nm (11 nm). It is also seen that silica core-gold nanoshells have smaller peak value at larger resonance wavelength than hollow core-gold nanoshells of the same dimensions when the shell thickness is thinner, while they have almost the same peak position and intensity when the shell thickness is thicker (Fig. 5).

3.3. Optimization

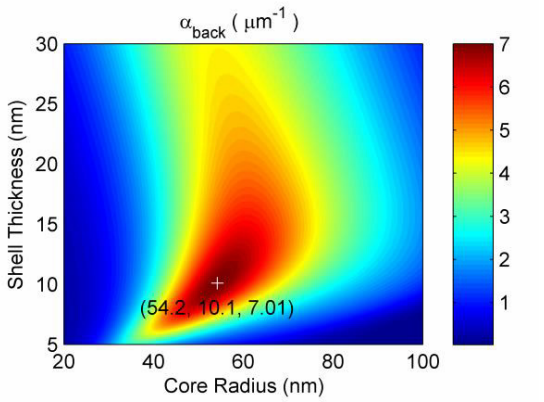

Fig. 6. Volume backscattering coefficient (α_{back}) of silica core-gold nanoshells at wavelength of 830 nm as a function of the core radius and shell thickness. The plus sign (+) is a point corresponding to the maximum α_{back}, and the numbers in the bracket are the values of optimal core radius, shell thickness, and α_{back}, respectively.

Table 1. Maximum volume backscattering coefficient ($\alpha_{back,max}$) and the corresponding optimal core radii ($R_{1,opt}$) and shell thicknesses (t_{opt}) of gold nanoshells with silica and hollow cores at four typical wavelengths.

	λ (nm)	$\alpha_{back,max}$ (μm^{-1})	$R_{1,opt}$ (nm)	t_{opt} (nm)
Silica core	830	7.01	54.2	10.1
	840	6.87	55.3	10.0
	900	6.03	62.1	9.4
	1310	2.71	110.0	8.0
Hollow core	830	7.23	54.8	8.7
	840	7.08	56.0	8.6
	900	6.21	62.7	8.1
	1310	2.76	110.3	7.0

From the results above, it can be known that by varying the dimensions of the core and shell, the backscattering resonance of gold nanoshells can be precisely and systematically varied over a broad region ranging from the visible to the near-infrared. Furthermore, the backscattering resonance of gold nanoshells can also be tuned to show the same peak position but different peak intensity with different size parameters. Therefore, one can design the dimensions of gold nanoshells at an incident wavelength in order to they have maximum backscattering for biological imaging applications. If the wavelength of the incident light is given, the optimal core radius and shell thickness of gold nanoshells can be easily obtained by maximizing the volume backscattering coefficient. In this paper, four typical wavelengths (830, 840, 900, and 1310 nm) commonly-used in biological imaging with OCT [15-

19] are considered. Fig. 6 shows the volume backscattering coefficient of silica core-gold nanoshells at wavelength of 830 nm as a function of the core radius and shell thickness. Clearly, the gold nanoshells possess maximum volume backscattering coefficient (7.01 μm^{-1}) when the core radius is 54.2 nm and shell thickness is 10.1 nm. Maximum volume backscattering coefficient and the corresponding optimal core radii and shell thicknesses of gold nanoshells with silica and hollow cores at four typical wavelengths (830, 840, 900, and 1310 nm) are shown in Table 1. For longer wavelength, the maximum volume backscattering coefficient becomes small, and the corresponding core radius is larger while the shell thickness is thinner. From the comparison of the optimized results for silica and hollow core-gold nanoshells, we can see that hollow core-gold nanoshells possess bigger maximum volume backscattering coefficient at the same wavelength, and they have a little bigger core radius and thinner shell thickness. At the optimal condition, hollow core-gold nanoshells with core radius of 54.8 nm and shell thickness of 8.7 nm have the maximum volume backscattering coefficient (7.23 μm^{-1}) at wavelength of 830 nm, which can be used as the most ideal contrast agents for biological imaging.

4. Conclusion

In summary, using Mie theory of a coated sphere with the size-dependent dielectric function of metal nanoparticles, we investigated the effects of the core material and particle size on the backscattering properties of gold nanoshells. By varying the core material and dimensions of the core and shell, the resonance wavelength can be precisely and systematically tuned over a broad region ranging from the visible to the near-infrared, and the resonance peak intensity can be changed. With the increase of the core radius, the resonance wavelength increases near-linearly, and the resonance peak intensity increases sharply first, then reaches maximum, and finally decreases. With the increase of the shell thickness, the resonance wavelength decreases sharply first, then reaches minimum, and finally increases, while the resonance peak intensity increases sharply first, then reaches maximum, and finally decreases. We obtained the optimal dimensions of gold nanoshells which possessing maximum backscattering at four typical wavelengths (830, 840, 900, and 1310 nm) frequently-used in biological imaging. It was found that silica core-gold nanoshells have maximum volume backscattering coefficient (7.01 μm^{-1}) at wavelength of 830 nm when the core radius is 54.2 nm and the shell thickness is 10.1 nm. For longer wavelength, the optimal core radius is larger while the optimal shell thickness is thinner, and the maximum volume backscattering coefficient become small. If the core material is changed from silica to vacuum, the gold nanoshells have bigger maximum volume backscattering coefficient with larger core radius and thinner shell thickness. The optimized gold nanoshells can be used as the ideal contrast agents for biological imaging. In this work we present an optimization idea for gold nanoshells. This straightforward idea can be directly applied to other metals with only the adjustment of the parameters in the size-dependent dielectric function. Additional particle geometries such as nanocages and nanorods, which have been commonly-used in biological imaging, can also be optimized by using numerical methods such as T-matrix, discrete dipole approximation (DDA), and finite-difference time-domain (FDTD).

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